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DOE/SC-ARM-22-002

Examining the Ice-Nucleating Particles from the Eastern North Atlantic (ExINP-ENA) Field Campaign Report

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February 2022

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Work supported by the U.S. Department of Energy,
Office of Science, Office of Biological and Environmental Research

Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
DOE	U.S. Department of Energy
ENA	Eastern North Atlantic
ExINP-ENA	Examining the Ice-Nucleating Particles from the Eastern North Atlantic
HPLC	high-performance liquid chromatography
INP	ice-nucleating particle
IOP	intensive operational period
IT	information technology
NOAA	National Oceanic and Atmospheric Administration
NSA	North Slope of Alaska
PI	principal investigator
PINE	Portable Ice Nucleation Experiment
SGP	Southern Great Plains
UPS	uninterruptible power supply
UTC	Coordinated Universal Time
WT-CRAFT	West Texas Cryogenic Refrigerator Applied to freezing Test

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1.0 Summary

The Examining the Ice-Nucleating Particles from the Eastern North Atlantic (ExINP-ENA) field campaign was conducted at the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility's Eastern North Atlantic (ENA) observatory on Graciosa Island, Azores (39.0916° N, 28.0257° W). The location of the ENA site is shown in Figure 1.

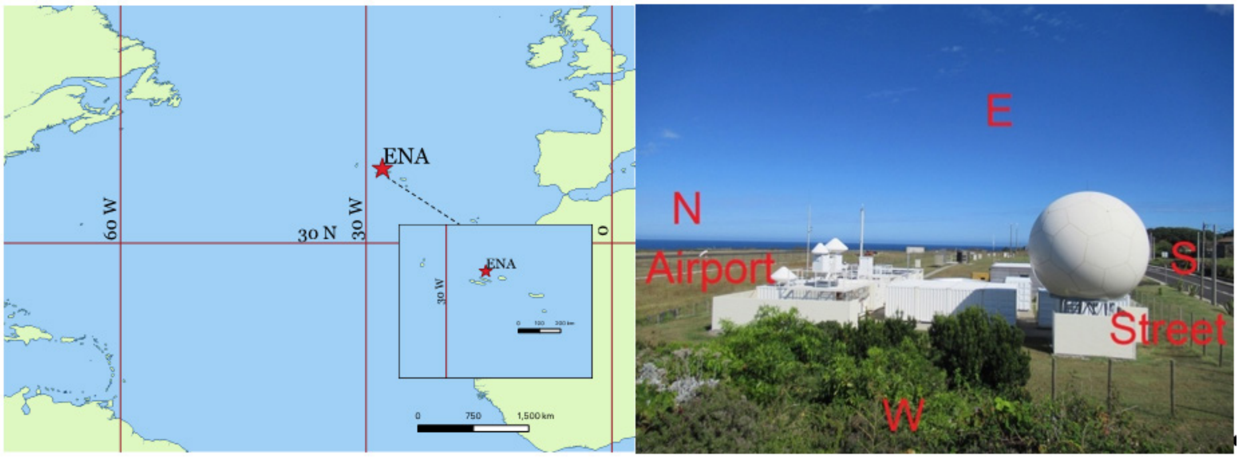

Figure 1. The ExINP-ENA campaign took place at the DOE ARM Eastern North Atlantic observatory, indicated on the map with a red star (inset shows the location of Graciosa Island within the Azores) (a). The sampling location on Graciosa Island is south of the airport and north of a street, as shown in panel (b).

The ExINP-ENA campaign began on October 1, 2020, and collected data until March 28, 2021, for a total of 179 days of data generated, with a 45-day intensive operational period (IOP) from October 10 to November 24. This campaign was funded by the DOE Office of Science Early Career Research Program through grant DE-SC001879. This grant contains funding for three field campaigns. The previous field campaign at the Southern Great Plains (SGP) site in Oklahoma, ExINP-SGP, was completed in 2020 (36.6073° N, 97.4876° W), and the upcoming field campaign at the North Slope of Alaska (NSA), ExINP-NSA, will take place from October 2021 at the National Oceanic and Atmospheric Administration (NOAA) Barrow Atmospheric Baseline Observatory (71.3230° N, 156.6114° W). The campaign aims included:

- Determining the concentration of ice-nucleating particles (INPs) active at temperatures spanning the range of heterogeneous freezing processes (from ≈-30 °C to 0 °C) using a combination of online and offline measurements
- Determining whether local meteorological conditions impacted INP concentration (N_{INP}) and/or ice nucleation temperature
- Using concurrent instrumentation to examine physicochemical properties of INPs as they relate to aerosol chemistry
- Determining if there is a relationship between INPs and cloud condensation nuclei (CCN) at the ENA site.

Current high-time-resolution INP data sets are lacking, and no previous studies have examined INPs at the ENA site, providing this campaign the opportunity to generate a unique and much-needed data set that can be used in collaboration with other groups to refine current Earth system models. As well, one of the stated goals of ARM is to advance understanding of aerosol-cloud ice interaction, which will be a direct result of this campaign. Current climate models poorly represent INPs, and the 15-minute-time-resolution data over several seasons generated during this campaign will provide an invaluable resource, especially combined with the data sets generated during ExINP-SGP and eventually ExINP-NSA. These data sets will allow for a greater understanding of ice nucleation processes as they may (or may not) relate to local meteorological processes and aerosol chemistry and will eventually help further understanding of the Earth's climate processes and energy balance.

This report includes N_{INP} measured with the Portable Ice Nucleation Experiment (PINE) chamber at the ENA site. The PINE chamber measures N_{INP} by sampling 10 L of ambient air into an aluminum expansion chamber, which then undergoes simulated adiabatic cooling and ice supersaturation condition as the pressure inside the chamber is lowered. The PINE chamber has a temperature uncertainty of approximately ±1.5 °C and a lower concentration detection limit of approximately 0.3 INP L^{-1}. Further details of the PINE chamber can be found in Möhler et al. (2021). Measurements made with the PINE chamber give a time resolution of approximately 15 minutes between each measurement, and the instrument is able to scan and collect data at multiple points between -30 °C and -15 °C over the course of approximately two hours.

During this campaign, the PINE chamber was housed in an air-conditioned container at the ENA site as shown in Figure 2. We employed a homemade quasi-laminar sampling inlet to sample ambient air for PINE and other sampling activities conducted in the container. Our sampling line particle loss test indicated negligible particle loss for 0.5-5 μm aerodynamic particle diameter (D_{p}) particles at the sampling inlet of PINE while a slight suppression of >8 μm as well as ≤0.5 μm D_{p} particles is observed (presumably due to gravitational loss and diffusional loss of particles, respectively).

This campaign proved the ability of the PINE chamber to collect data with minimal in-person interaction with the instrument. The instrument setup was performed by a collaborator from Karlsruhe Institute of Technology due to travel restrictions. The instrument was then remotely controlled using a custom-designed LabView program (control panel shown in Hiranuma and Vepuri 2020) for the entire time period other than during instrument reset times, when onsite technicians were available to restart the instrument and reset the computer hardware.

Figure 2. A 5.5-m-height (0.1 m diameter) cylinder-shaped sampling stack inlet was mounted to the instrument container, allowing an intake of particle-laden air by the PINE chamber and other instruments inside.

The technicians also assisted in the collection of filter samples that were later analyzed using an offline freezing technique, called West Texas Cryogenic Refrigerator Applied to freezing Test (WT-CRAFT), described in Hiranuma et al. (2021) and Vepuri et al. (2021). Briefly, samples were collected onto polycarbonate Whatman Nuclepore 47-mm filters (0.2-µm-diameter pore size) for three to four days. Sampled air volume was estimated based on the sampling airflow and period for each filter. Filters were then stored in sterile Petri dishes at -20 °C (other than during transportation between Graciosa Island and Texas, when they were stored at ambient temperatures) prior to analysis, which occurred no more than one year after collection. Filters were washed in HPLC-grade water (Sigma Aldrich) to resuspend aerosol samples. The ice nucleation ability of these aerosols was then tested by plating on a machined aluminum plate inside a cryocooler that cooled the sample at a rate of 1 °C per minute. Based on the number of droplets frozen, the N_{INP} value was calculated using an equation based on calculations given in Vali (1971). The presence of proteinaceous and/or heat-sensitive aerosols was examined by comparing freezing spectra from each filter before and after the aerosols were heated at 100°C. A reduction in INPs was attributed to heat-sensitive, likely primarily proteinaceous, aerosols.

2.0 Results

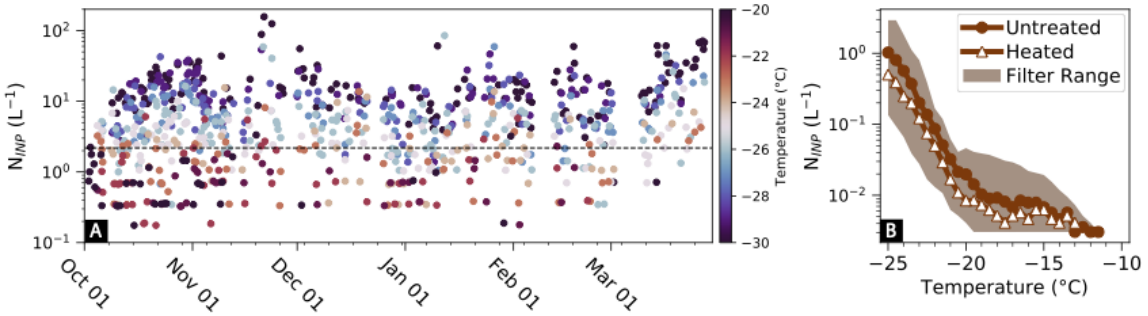

Figure 3. (a) Six-hour-averaged INP concentration, NINP, measured with the PINE chamber at ENA from October 2020 to March 2021 (UTC). The dashed line represents the average NINP at -25°C reported by past studies in the Atlantic Ocean (see text for a full list of references). (b) Median NINP measured from polycarbonate filters taken at ENA during meteorological autumn (filters from October 5 to November 30, 2020). The shaded area represents the minimum and maximum NINP measured from untreated filters at each 0.5°C temperature interval.

Figure 3a shows the PINE data taken during the study (data points represent 6-hour averages). All timestamps are based on Coordinated Universal Time (UTC). The N_{INP} value over the entire study period ranged from 0 to 105.5 INP L^{-1}, with the maximum N_{INP} measured on the afternoon of October 30 at -30 °C. There were multiple data points with 0 INPs measured. The lowest non-zero measurement was made in the early morning on November 5, with 0.07 INP L^{-1} measured at -22 °C. As there have not been any prior measurements of INPs at ENA, finding comparable studies is difficult. To overcome this, median values measured with the PINE chamber at ENA can be compared to the range of values of INPs measured at sites with influence from the Atlantic Ocean (Córdoba et al. 2021, DeMott et al. 2016, Flyger and Heidam 1978, Gong et al. 2019, 2020, Ladino et al. 2019, McCluskey et al. 2018, Rosinski et al. 1988, Sanchez-Marroquin et al. 2020, Sanchez-Marroquin, et al. 2021, Welti et al. 2018, 2020). Although not fully comparable, as many of these studies are from drastically different latitudes, our measurements are well within the range of Atlantic marine INPs measured by previous studies using both online and offline measurements.

Figure 3b shows median N_{INP} determined from samples collected on filters and analyzed using an offline method. Median N_{INP} values from filters range from 1.03 L^{-1} at -25 °C (the coldest temperature measured with WT-CRAFT) to 0.003 INP L^{-1} at -13 °C, with the concentration generally increasing with decreasing temperature in both data sets. Although some filter samples show a lower N_{INP} than that measured by the PINE chamber at the same temperature, this is to be expected and is not an area of concern. Several factors may drive this discrepancy. First, the technique employed with the WT-CRAFT instrument is only capable of measuring the concentration of INPs driving immersion freezing processes, while the PINE chamber can measure the concentration of ice crystals nucleated by both immersion and deposition freezing processes under ice supersaturation, yet water subsaturation, condition (Möhler et al. 2021). As well, while filter samples were held at -20 °C until processing, it is possible that some organic INPs were lost to degradation over the course of the storage time. In contrast, INPs measured with the PINE chamber are not stored, heated, or treated in any way and are measured immediately following sampling. This leads to a potential for higher measured PINE N_{INP} when compared with N_{INP} measured at the same temperature from samples collected on filters.

Filter samples were also heat-treated at 100 °C for 20 minutes. Heat treatment was applied by sealing the sample in a sterile sample tube and placing this tube in a beaker full of boiling water for 20 minutes. Although we cannot definitively assert that this method removes only proteinaceous INPs (Daily et al. 2021), we are confident that this method results in the near-complete removal of proteinaceous INPs, as irreversible protein denaturation begins at only 40 °C (Hogg 2013). Some of the previous studies (conducted in a different location) employing this method have seen large reductions in N_{INP} after heat-treating samples (e.g., Hill et al. 2016). The N_{INP} values measured from filter samples decreased with heat-treatment for 7.5 – 57.3%, with the greatest proportional heat sensitivity seen at -20 °C. The average reduction in N_{INP} after heat treating was 37.1%. At -20 °C the average N_{INP} in the untreated sample was 1.97×10^{-2} L^{-1}, while the average concentration in the heated sample decreased to 8.42×10^{-3} INP L^{-1}. This suggests that the INPs are dominated by non-heat-labile material. Although the exact nature of this material is unknown, illumination of its composition represents an excellent goal for future studies in the region.

To illuminate patterns in N_{INP}, measurements of N_{INP} made by the PINE chamber were compared with several different measurements made by collocated instruments maintained as part of the ongoing measurements made at ENA. There was no relationship between N_{INP} and any local meteorological data, including precipitation, wind speed, temperature, cloud base height, or wind direction. However, this is not unexpected as there is potential for INPs to travel to ENA from many kilometers away. This lack of correlation suggests that local weather processes are not a major source of INPs. As well, total aerosol concentrations did not correlate with N_{INP}. At other sites, INPs are by no means a majority of the aerosol population, and may only compose as few as one ice-nucleation active particle in a million aerosol particles.

The INP data set generated by this campaign represents a one-of-a-kind data set for this location, as no previous study at the ENA site has included both online and offline INP measurements, let alone collected data for more than 45 days. Our 45-day data set includes N_{INP} measured at a time resolution as high as every 15 minutes, and will provide valuable information for climate modelers who wish to link INPs to other climate processes. Future work on this project will include the publication of a paper regarding PINE, filter, and other aerosol physicochemical data from ENA and SGP (to be submitted Q1 2022), a continuation of laboratory examination of sample chemistry through offline microspectroscopy, and eventual publication of a paper comparing all of the data collected through ExINP field campaigns, including SGP, ENA, and NSA sites (submission date to be determined). Future campaigns at ENA would allow for the comparison of interannual variability in INP and CCN populations, as well as providing stronger information on seasonal variability by removing single-year anomalies. Increased aerosol chemistry data would be useful to determine if there is a specific aerosol composition that contains a high N_{INP}.

3.0 Publications and References

1. A multi-site comparison of aerosol properties, including ice-nucleating particles, measured during autumn field campaigns with online and offline techniques

   Wilbourn, EK, L Lacher, M Mazzola, O Möhler, and N Hiranuma
   Submitted, European Geosciences Union Meeting, 2022

2. Ice nucleating particle concentrations from an arctic and a temperate site with marine-dominant aerosol sources

   Wilbourn, EK, N Hiranuma, M Mazzola, R Traversi, L Lacher, Y Hou, C Guerrero, S Alrimaly, and O Möhler
   Poster, American Geophysical Union Fall Meeting, 2021

3. Evaluating ice-nucleating particles from E3SM model in the Southern Ocean and Eastern North Atlantic Ocean

   Raman, A, EK Wilbourn, PJ DeMott, T Hill, N Hiranuma, and SM Burrows
   Presentation, American Geophysical Union Fall Meeting, 2021

4. Comparing online and offline measurements of ice-nucleating particles from two autumn field campaigns

   Wilbourn, EK, L Lacher, HSK Vepuri, J Nadolny, O Möhler, and N Hiranuma
   Presentation, American Association for Aerosol Research Meeting, 2021

5. A comparison of aerosol particle sources and ice-nucleating particle properties from the Eastern North Atlantic and U.S. Southern Great Plains

   Wilbourn, EK, N Hiranuma, HSK Vepuri, L Lacher, J Nadolny, and O Möhler
   Presentation, European Aerosol Conference 2021

6. An abundance of ice-nucleating particles in the Atlantic sector of the Arctic and the mid-latitude sites
   Hiranuma, N, HSK Vepuri, and EK Wilbourn
   Presentation, TAMU-ATMO seminar, 2021

7. A new instrument for semi-autonomous measurements of atmospheric ice-nucleating particles: the Portable Ice Nucleation Experiment (PINE)

   Murray, BJ, KS Carslaw, PR Field, T Storelvmo, N Hiranuma, L Lacher, MP Adams, A Hobl, F Vogel, and O Möhler
   Poster, WMO Global Atmosphere Watch Programme symposium 2021, 2021.

8. Ice-nucleating particle concentration measurements from the ARM Eastern North Atlantic and Southern Great Plains observatories

   Hiranuma, N, EK Wilbourn, HSK Vepuri, L Lacher, J Nadolny, and O Möhler
   Presentation, USDOE ARM PI Meeting, 2021

9. Remotely controlled ice-nucleating particle measurements from the Eastern North Atlantic during autumn and winter

   Wilbourn, EK, N Hiranuma, L Lacher, J Nadolny, and O Möhler
   Presentation, European Geophysical Union Annual Meeting, 2021

10. Characterization and first applications of the Portable Ice Nucleation Experiment (PINE)
    Lacher, L, F Vogel, J Nadolny, R Ullrich, N Büttner, M Adams, C Boffo, T Pfeuffer, A Hobl, M Weiß, HSK Vepuri, EK Wilbourn, N Hiranuma, BJ Murray, and O Möhler
    Presentation, 10th virtual INP Colloquium, 2021

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5.0 Lessons Learned

1. Pre-campaign documentation (Instrument Support Requests, Site Visit Requests, Pre-IOP planning and meetings): is the process clear or confusing?

   The process was straightforward and intuitive. Heath Powers, John Archuleta, and other ENA admins and technicians have been communicative and helpful. The PI's team and ENA officers had several meaningful remote meetings to plan the campaign logistics prior to the site visit of Larissa Lacher.

2. Site Facilities, SGP Staffing and Assistance, Resources. (Were they adequate? How did they contribute to the outcome of the campaign?)

   Yes. All support was adequate and absolutely above average. With remote guidance of the PI, onsite ENA staff routinely performed the onsite device maintenance for the PI's team. Thus, they very positively contributed to the success of this campaign. They shared the project updates with the PI weekly (otherwise upon a request of the PI's team) and understood the importance of this project. The PI's team acknowledges (including but not limited to) Tercio Silva , Bruno Cunha, Carlos Sousa, Pawel Lech, Hannah Frances Ransom, John Archuleta, Heath Powers, and Karen Caporaletti. Our half-year campaign could not be completed without their support.

3. Safety, Training and Rollout. (Was the safety briefing adequate and beneficial? Was safety a priority? Were there any incidents or near misses? Were severe weather notices acceptable? Were there adequate storm shelters? What could be improved upon?)

   Under the current pandemic circumstance, safety was indeed prioritized professionally at ENA. The COVID safety protocol for Lacher was well instructed, constructive, and intuitive to follow. Although no severe weather conditions were seen while Larissa Lacher was at ENA, the notification method was appropriate.

4. Expectations and Objectives.(Were the expectations and objectives achieved?)

   Yes. We achieved our objectives with a contingency plan (refer to the response in Q7).

5. Project Definition, Planning, and Control throughout the campaign (Was there a project plan? Was it used? Was it realistic?)

   N/A

6. Organization –Meetings, communication, information sharing (Who managed most of the communication within the IOP? Did it work?)

   All went well. The ENA onsite staff (especially Tercio Silva , Bruno Cunha, Carlos Sousa) and the PI's team communicated frequently through virtual meetings throughout the campaign. Offsite support of the ENA admins, an IT technician (Pawel A. Lech), as well as the ARM-ENA logistics team (Hannah Frances Ransom and Karen Caporaletti), were always helpful and appreciated, too.

7. Risk and scope change throughout the project.(Did the project change significantly? How was this managed?)

   Yes. Due to travel restrictions, Hiranuma and Wilbourn could not make it to the ENA site and gave up some of the planned activities (e.g., suspension sampling). However, as a contingency plan, we carried out longer team filter sampling and some other associated onsite activities with support of ENA technicians. In the end, the project was completed as the PI planned and proposed plus we collected an unexpectedly extended data set.

8. Campaign Wrap-up (Was there adequate manpower to take down equipment, load it, and leave the location that you occupied in an acceptable manner?) If not, please give suggestions.

   The PI's team appreciates the ENA technicians for tearing down our instruments through our remote instruction. Logistics afterward went smoothly too. All instruments have been safely transported to the next destination, North Slope of Alaska site, and they are currently running there.

9. Overall IOP success

   100/100 – mission complete.

10. What went well, and why?

    Basically everything. The ENA staff are professional. Their attention, consideration, and ability for problem-solving made it possible to process any requests that the PI's team made promptly.

11. What went badly, and why?

    Nothing.

12. What could have been done differently, with hindsight?

    Due to stormy weather conditions, the network and power conditions were often unstable. Consequently, the PI's team sometimes lost remote control of our instruments, which caused some minor issues, such as prolonged maintenance and data gap. These infrastructures are crucial for any remote campaigns. Therefore, stable network connection and full-time backup power (via uninterruptable power supply [UPS]) would be a nice addition to the ENA site in the future.

13. What are lessons for future IOPs?

    The PI's team had a very meaningful outcome and positive experience from the ExINP-ENA campaign. Building relationships and trust with the ARM site managers and onsite technicians is one of the most important aspects of any ARM project. Thus, the PI considers that the capability to build teamwork for projects is key for success in ARM IOPs.

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